

Classical spin-liquid on the maximally frustrated honeycomb lattice

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We show that the honeycomb Heisenberg antiferromagnet with $J_1/2 = J_2 = J_3$, where $J_{1/2/3}$ are first-, second- and third-neighbour couplings respectively, forms a classical spin liquid with pinch-point singularities in the structure factor at the Brillouin zone corners. Upon dilution with non-magnetic ions, fractionalised degrees of freedom carrying $1/3$ of the free moment emerge. Their effective description in the limit of low-temperature is that of spins randomly located on a triangular lattice, with a frustrated interaction of long-ranged logarithmic form. The XY version of this magnet exhibits nematic thermal order by disorder, which comes with a clear experimental diagnostic.

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Motivation.—The honeycomb lattice has – somewhat belatedly – become one of the prime hunting grounds for spin liquids (SL) in $d = 2$ [1], in addition to the kagome and the $J_1 - J_2$ square lattice Heisenberg models, which have been the focus of much attention over decades, continuing until today. In both these latter cases [2–11], confidence in the existence of a quantum SL state for $S = 1/2$ magnets has ebbed and flowed, while the classical (large-spin) versions evade liquidity by exhibiting – rather interesting – forms of order by disorder [12–20].

The richness of magnetic models on the honeycomb lattice – bipartite, like the square lattice – has therefore come as somewhat of a surprise. Initially emulating its brethren by appearing to support a quantum SL in a Hubbard model [21], it has been attracting attention in the context of the fractionalised phases of the Kitaev honeycomb model [22], exhibiting highly unusual exactly soluble quantum SL phases. Particular impetus arose from the suggestion that the Kitaev Hamiltonian may describe the materials $\{Na, Li\}_2IrO_3$, provided a Heisenberg term is added [23–25].

In fact, detailed studies of these materials suggest that further nearest neighbor terms play an important role in explaining spiral ordering at low temperatures [26], and one of the models studied in some detail is the $J_1 - J_2 - J_3$ Heisenberg model, which had already been subject to considerable earlier attention [27–30]. In determining the Hamiltonian appropriate to these materials, it has turned out to be instructive to consider their response to disorder [31].

Here, we identify and study in detail an unusual, hitherto overlooked, classical SL state on the honeycomb lattice, associated with the (known) degeneracy point $J_1/2 = J_2 = J_3$ of the Heisenberg model on the honeycomb lattice. It exhibits remarkable new features. These arise from the fact that the dual lattice, as well as the underlying Bravais lattice, is the tripartite triangular lattice. They include pinch points in the structure factor at the zone corner wavevector \mathbf{Q} (which distinguishes be-

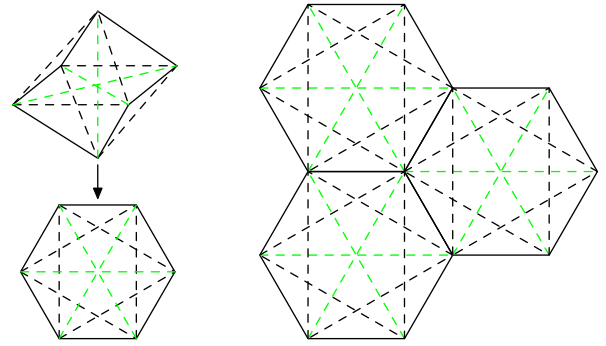


FIG. 1: (color online). Projection of the octahedron into the hexagon and the $J_1 - J_2 - J_3$ model on the honeycomb lattice. The J_3 interactions are differentiated with colors.

tween the three sublattices), as well as novel disorder effects whereby, upon dilution, fractionalised moments carrying *one third* of the microscopic spin moment appear. These fractionalized moments interact via a frustrated, sublattice-dependent, long range interaction in the limit of low temperature, T .

This model is further remarkable as it can be thought of the first realisation of a SL in $d = 2$ of *edge-sharing* simplices, which here take the form of octahedra. In addition, its XY version does exhibit nematic order by disorder, which turns out to be straightforwardly detectable in neutron scattering through the appearance of peaks in the structure factor.

The remainder of this paper is organised as follows. We first introduce the model and derive and describe its SL, for which we formulate a novel low-energy description. We then study its behaviour under dilution. All our analytical predictions are supported by Monte Carlo (MC) simulations of the microscopic Hamiltonian. We close with an outlook, in which we argue that this model is quite natural, as (i) the degeneracy point corresponds to a reasonably natural set of parameter values; and (ii) we expect the SL to fan out as T is increased, at the

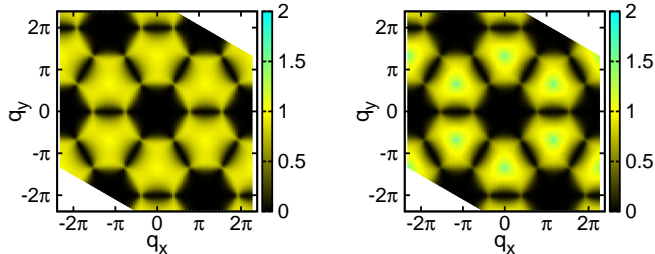


FIG. 2: (color online). Structure factor as obtained in Monte Carlo simulations of the pristine Heisenberg (left) and XY (right) systems. Both results correspond to $N = 1800$ spins at $T/J = 0.01$.

expense of adjacent phases exhibiting lower entropies.

Model.—The Hamiltonian for classical $O(n)$ spins \vec{S}_i of unit length on sites i of the honeycomb lattice reads:

$$\begin{aligned} H &= J_1 \sum_{\langle i,j \rangle} \vec{S}_i \cdot \vec{S}_j + J_2 \sum_{\langle\langle i,k \rangle\rangle} \vec{S}_i \cdot \vec{S}_k + J_3 \sum_{\langle\langle\langle i,l \rangle\rangle\rangle} \vec{S}_i \cdot \vec{S}_l \\ &= \frac{J}{2} \sum_{\alpha} (\vec{S}_{\alpha}^{\alpha})^2 + \text{const.}, \end{aligned} \quad (1)$$

where $\langle i,j \rangle$, $\langle\langle i,k \rangle\rangle$ and $\langle\langle\langle i,l \rangle\rangle\rangle$ refer respectively to first, second and third nearest neighbour pairs, while the second line follows from fixing $J_1/2 = J_2 = J_3$.

This form shows that each and any configuration where each hexagon, labelled by α , has vanishing total spin, $\vec{S}_{\alpha}^{\alpha} = 0$, is a ground state. Such a rewriting is often helpful for geometrically frustrated lattices. It is most often used for ‘corner-sharing’ structure of elementary simplices [19, 20], examples being pyrochlore (corner-sharing tetrahedra) or kagome (corner-sharing triangles) lattices. It immediately allows to estimate the dimensionality of the ground state manifold, F . This proceeds by subtracting the number of constraints, K , imposed by Eq. 1, from the total number of degrees of freedom, D , of the spin system.

For a system of n -component spins with N such simplices, and each spin part of b simplices, $D = q(n-1)/b$ per simplex, where the number of spins in a simplex $q = 3, 4, 6$ for triangle, tetrahedron, octahedron respectively. Each simplex imposes $K = n$ constraints, as each component of its total spin must vanish. Hence,

$$F = \frac{q(n-1)}{b} - n. \quad (2)$$

To maximize F , and hence enhance the chance of finding a SL [19, 20], one thus should minimize b , or maximise n and q . Indeed, b is minimal for corner-sharing arrangements, and $q = 4$, $n = 3$ result in the well-established classical SL on the pyrochlore lattice. Triangle-based lattices (kagome has $q = 3$) need higher, $n \geq 4$, component spins for a similar SL to arise [32].

The $J_1 - J_2$ model on the square lattice with $J_2 = J_1/2$ can be thought of as edge-sharing tetrahedra, with a large $q = 4$; it does not support $F > 0$ for any n . Indeed, no such Heisenberg model with $F > 0$ has been identified for edge-sharing simplices at all so far.

However, from Eq. 2, $F = 1$ for $q = 6$ and $b = 3$, which corresponds to the frustration point of the honeycomb lattice, Eq. 1! It can be thought of as edge-sharing octahedra (Fig. 1), and thus presents the first instance of a possible SL on an edge-sharing lattice. It is also the first with $b > 2$, a fact with significant consequences as we explore in detail below.

Before we do this, we demonstrate that this model does indeed exhibit a SL with algebraic correlations for $T \rightarrow 0$, by analytically evaluating the correlations for soft spins (equivalently, in the large- n or self-consistent Gaussian approximation [33]) and comparing the result to classical MC simulations. These yield a $T = 0$ structure factor presenting *pinch points*, the defining characteristic of such algebraic SLs [34]. Somewhat unusually, in this case the pinch points are located at the corners of the Brillouin zone.

From our MC simulations for Heisenberg and XY spins, we plot structure factor and specific heat on Figs. 2, 3. The MC simulations employ a combination of heat-bath and microcanonical moves as well as parallel tempering moves. The structure factor from MC simulation of Heisenberg spins agrees with the analytical prediction for soft spins. By contrast, for $n = 2$, the corresponding XY model, low temperature peaks develop in addition to the pinch points. This is an instance of nematic (collinear) order by disorder, as is readily verified by constraint counting [19, 20]. We note in passing that the appearance of these peaks provides an unusually direct signature of collinear ordering.

This interpretation is confirmed by a low- T specific heat of $c = 0.375k_B$ per spin, reduced from the value of $c = bnk_B/2q$ expected from equipartition in the absence of order by disorder [15–19], as is found in the Heisenberg magnet with $c = 0.75k_B$.

The existence of pinch points in the Heisenberg case comes as somewhat of a surprise given the non-bipartite nature of the dual triangular lattice. In the corresponding corner-sharing models, the bipartiteness of the dual lattice (square, honeycomb or diamond lattice) is a crucial ingredient for such pinch points [34]. Indeed, in work close in spirit to the present one, on bosons on a honeycomb and the dual triangular lattice [35], one finds an Ising emergent gauge field implying the absence of pinch points. The way this issue resolves itself in the present case is quite interesting: First, the pinch points are located at the Brillouin zone corners, corresponding to a three-sublattice wave vector \mathbf{Q} . Second, the low-energy description is naturally expressed in terms of a vector field that captures slow modulations near wavevector- \mathbf{Q} , reminiscent of the two dimensional height field acting as

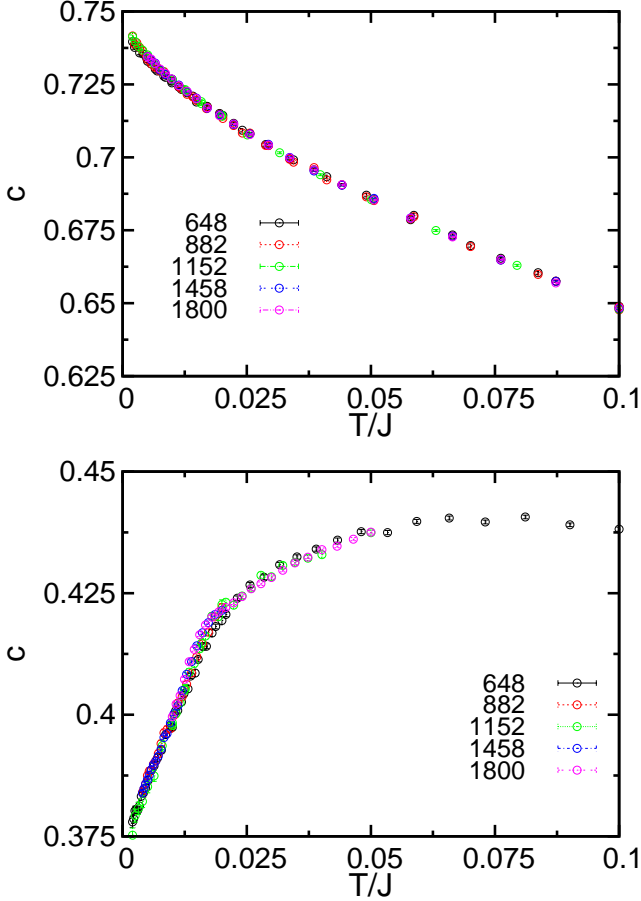


FIG. 3: (color online). Specific heat obtained in Monte Carlo simulations of the pristine Heisenberg ($n = 3$, top) and XY ($n = 2$, bottom) systems, with $c = \frac{3}{8}k_B < \frac{b\eta}{2q}k_B$ indicating nematic order by disorder for the XY case.

an emergent $U(1)$ gauge field [34]. In detail, this proceeds as follows.

Consider an A-sublattice (B-sublattice) site \vec{r}_A (\vec{r}_B) of the honeycomb lattice, which sits at the center of an “up-pointing” (“down-pointing”) triangle comprising dual lattice points \vec{R}_a , \vec{R}_b and \vec{R}_c belonging to the three sublattices of the tripartite dual triangular lattice. One writes the corresponding $O(n)$ spins $\vec{S}_{\vec{r}}$ in terms of $\vec{\zeta}_{\vec{R}}$ and $\vec{\tau}_{\vec{R}}$, two $O(n)$ vector fields on the dual triangular lattice.

$$\vec{S}_{\vec{r}_A} = \sum_{\alpha=a,b,c} (\vec{\tau}_{\vec{R}_\alpha} + \vec{\zeta}_{\vec{R}_\alpha}), \quad \vec{S}_{\vec{r}_B} = \sum_{\alpha=a,b,c} (\vec{\tau}_{\vec{R}_\alpha} - \vec{\zeta}_{\vec{R}_\alpha}).$$

In the self-consistent Gaussian approximation, the partition function for the Hamiltonian Eq. 1 can be written as a product of $\vec{\zeta}$ and $\vec{\tau}$ partition functions, with actions

$$\mathcal{S}_{\vec{\zeta}} = F_1(\{\vec{\zeta}\}), \quad \mathcal{S}_{\vec{\tau}} = F_1(\{\vec{\tau}\}) + F_2(\{\vec{\tau}\}), \quad (3)$$

where

$$F_1(\{\vec{v}\}) = \frac{\rho}{2} \sum_{\vec{r}} (\vec{v}_{\vec{R}_a(\vec{r})} + \vec{v}_{\vec{R}_b(\vec{r})} + \vec{v}_{\vec{R}_c(\vec{r})})^2 \quad (4)$$

for $\{\vec{v}\} = \{\vec{\zeta}\}$ or $\{\vec{\tau}\}$, and

$$F_2(\{\vec{\tau}\}) = \frac{\beta J}{2} \sum_{\vec{R}} (6\vec{\tau}_{\vec{R}} + 2 \sum_{\vec{R}_n \in \partial \vec{R}} \vec{\tau}_{\vec{R}_n})^2 \quad (5)$$

here, $\vec{R}_n \in \partial \vec{R}$ denotes the six dual triangular lattice sites \vec{R}_n that are nearest neighbours of the dual triangular lattice site \vec{R} . The stiffness constant ρ is adjusted to yield $\langle \vec{S}_{\vec{r}}^2 \rangle = 1$.

This action implies that $\vec{\zeta}$ encodes the $T = 0$ fluctuations of the classical SL, while $\vec{\tau}$ captures thermal fluctuations. The $T \rightarrow 0$ limit is thus characterized by a particularly simple action in which the $\vec{\tau}$ fields do not contribute. This action, as well as the expressions for the physical spins \vec{S} , are both invariant under $\vec{\zeta}(\vec{R}) \rightarrow \vec{\zeta}(\vec{R}) + \text{Re}(\vec{\chi} \exp(2\pi i \vec{Q} \cdot \vec{R}))$ for any $\vec{\chi}$.

Dilution Effects.—The ground states of SLs often are less revealing of their topological nature than their excitations. An elegant way to visualise the latter as effectively a ground state property is to introduce disorder which then nucleates excitations. In SLs, this is perhaps most easily done by replacing some of the magnetic ions with non-magnetic ones. For classical SLs, this dilution problem has been studied in some detail both experimentally [36–39], and theoretically [40–43]. In particular, for the cases of SCGO, the checkerboard and the pyrochlore lattices, it was found that fractional impurity moments carrying one half of the moment of a free spin arise as a cooperative phenomenon. These so-called orphan spins occur when all but one of the spins of a simplex are replaced – so that the total spin of that simplex (see Eq. 1) can no longer possibly vanish.

These orphans turn out to provide a number of signatures of the new structure of the honeycomb SL. First of all, they directly reflect the fact that we have $b = 3$ *edge-sharing* octahedra meeting in each site – the fractional impurity moment is not one half but *one third* of that of a free spin! This is displayed in Fig. 4 (top panel) where a calculation based on a hybrid hard-soft spin theory [42, 43] is compared with numerical results for the local susceptibility. This is, to our knowledge, the first instance of fractionalisation into three items in a classical spin model.

Interactions between these orphans are entropic in nature and take the form of an effective Heisenberg exchange J_{eff} . They are mediated by the bulk SL, and hence reflect the structure of the latter. In the classical SLs known so far, these effective interactions can be written in a form which is uniformly antiferromagnetic [44]. Here, this is not possible: We now find that these interactions are antiferromagnetic / ferromagnetic for orphans residing on the same / different sublattice of the dual triangular lattice, respectively, with the antiferromagnetic interactions being twice as strong as the ferromagnetic ones. This intricate structure in the effective exchange couplings follows from our field theory,

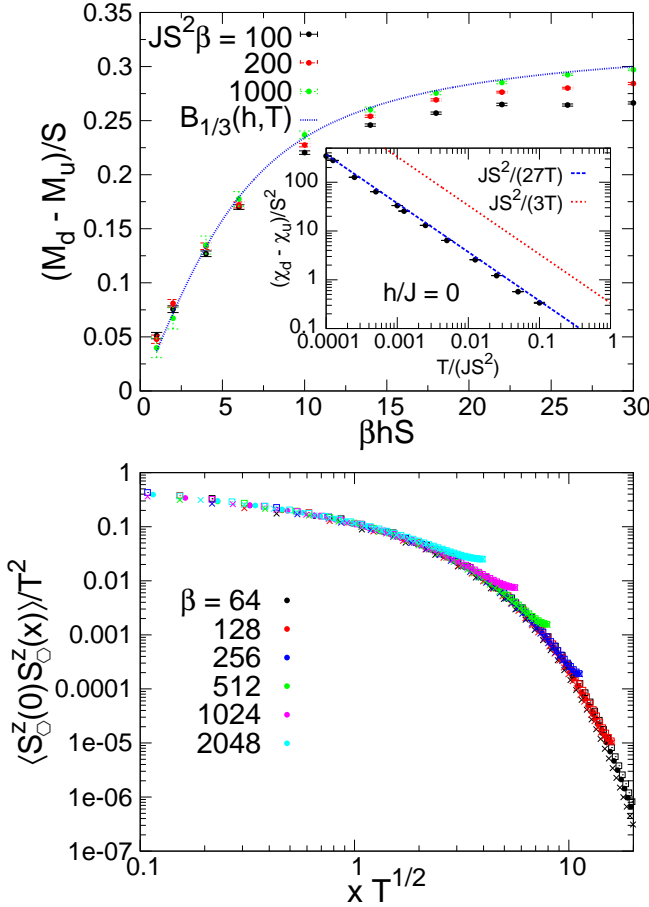


FIG. 4: (color online). **Top:** ‘Impurity magnetization’, defined as the difference of total magnetization in the diluted and undiluted systems, as observed in MC simulations of the model. The solid curve corresponds to the theoretical prediction for a free spin $S/3$ in a field h , i.e., the Langevin function $\mathcal{B}_{S/3}(h, T)$. The inset shows the ‘impurity susceptibility’ at zero external field, consistent with a Curie law for fractionalized spins $S/3$. **Bottom:** Testing the scaling prediction for the charge correlations on a finite lattice of linear size $L = 210$ using the expression for correlations within the soft spin approach. Crucially, correlations between sites on the same sublattice have been multiplied by an extra scaling factor of $-1/2$.

which relates these entropic interactions to a bulk property of the pristine spin liquid, namely the correlations between the thermally excited net spins \vec{S}_O (Eq. 1):

$$\beta J_{\text{eff}} \approx \frac{-\langle \vec{S}_{O, \vec{r}_1} \cdot \vec{S}_{O, \vec{r}_2} \rangle}{\langle \vec{S}_{O, \vec{r}} \cdot \vec{S}_{O, \vec{r}} \rangle^2}, \quad (6)$$

For low T and large distances $|\vec{r}_1 - \vec{r}_2| \gg a$, where a is the lattice spacing, this gives a scaling form:

$$\beta J_{\text{eff}} = \eta(\vec{r}_1, \vec{r}_2) \mathcal{F}((\vec{r}_1 - \vec{r}_2) \sqrt{T}) \quad (7)$$

$$\stackrel{T \rightarrow 0}{=} \frac{1}{2\pi} \eta(\vec{r}_1, \vec{r}_2) \log(\vec{r}_1 - \vec{r}_2), \quad (8)$$

where $\eta = +1$ ($\eta = -1/2$) if the orphans are on the same (different) sublattices of the dual triangular lattice. This is verified using the analytical large- n result for a finite lattice, Fig. 4. In the limit $T \rightarrow 0$, βJ_{eff} exhibits a long-ranged logarithmic form.

Outlook.—Our model, notwithstanding its simplicity, displays a plethora of phenomena of current interest; the unusual emergent $\vec{\tau}$ fields and the new fractionalized behavior of $1/3$ for the impurity spin moments show that even in a classical setting, these nontrivial phenomena, up to now apparently constrained to the quantum realm, can emerge. The particular new frustrated logarithmic interactions between the impurity moments are as yet unstudied, and will possibly lead to a spin glass, unlike in the bipartite cases [44].

As for realisations, the $2 : 1 : 1$ ratio of exchange interactions is natural if exchange is via an ion on the hexagon center with no angular dependence, as the nearest neighbors bonds are part of two hexagons. Known experimental values are encouragingly nearby, being close to $2 : 1.6 : 1.6$ [24]. Hence direct observation of these phenomena might be possible, the main obstacle perhaps being finite quantum fluctuations for $S = 1/2$. Quite generally, at finite T , the classical SL behavior will be favoured over competing phases on account of its large entropy, and in particular fan out from the degeneracy point. We hope that this work will incite further investigation on appropriate honeycomb materials.

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